Supporting Information

Experimental section

Materials

Kevlar 49 yarns were purchased from Dupont and used after dried at 50 °C in vacuum for 12 h. Dimethyl sulfoxide (DMSO), potassium hydroxide (KOH), polyvinyl alcohol (PVA), aniline monomer, and ammonium persulphate (APS), 12M (mol/L) HCl were purchased from Sinopharm Chemical Reagent Co. Ltd. All reagents were of analytical grade and used as received without further purification.

Preparation of ANFs-PVA Hydrogels

Aramid nanofibers (ANFs)/DMSO dispersion were prepared according to the method reported in the reference.[1] Typically, bulk Kevlar 49 yarns (6.0 g) and KOH (9.0 g) were added to DMSO (300 mL) and stirred vigorously at room temperature for 3 weeks to obtain 20 mg/mL ANFs/DMSO dispersion. Then, 5 mL of ANFs/DMSO dispersion was mixed with an equal volume of a 100 mg/mL PVA (Mw 145000–165000 a.u.) solution in DMSO. The mixture system was cast into a mold and immersed in water to form ANFs/PVA hydrogels with the desired shape.

Preparation of APP Hydrogels

The ANFs-PVA hydrogel with a size of 1 × 3 × 0.03 cm^3 was immersed in 5 mL of 1 M HCl aqueous solution containing different initial concentrations of aniline monomer, and kept at 0 ℃ for 12 h. APS (mole ratio to aniline = 1:1, dissolved in 5 mL of 1 M HCl), pre-cooled to 0 ℃, was poured into the above mixture, and the mixture was polymerized in situ at 0 ℃. Finally, the composite hydrogel was thoroughly washed by HCl aqueous solution, ethanol and deionized water, respectively. According to the loading amount of polyaniline of 9.2 wt%, 14.7 wt%, 18.1 wt%, and 20.6 wt% (relative weight ratio of PANI to ANFs-PVA-PANI (APP) hydrogels), APP hydrogels (85% water content) were labeled as APP-0, APP-1, APP-2, APP-3 and APP-4, respectively.

Assembling of Strain Sensors Utilizing APP Hydrogels

To obtain the strain sensor, the excess water on the surface of APP-3 hydrogel (1 × 3 ×

0.05 cm^3) was absorbed with filter paper, then two copper plates attached with conductive silver paste were connected to the both ends of the hydrogel (Figure 1).

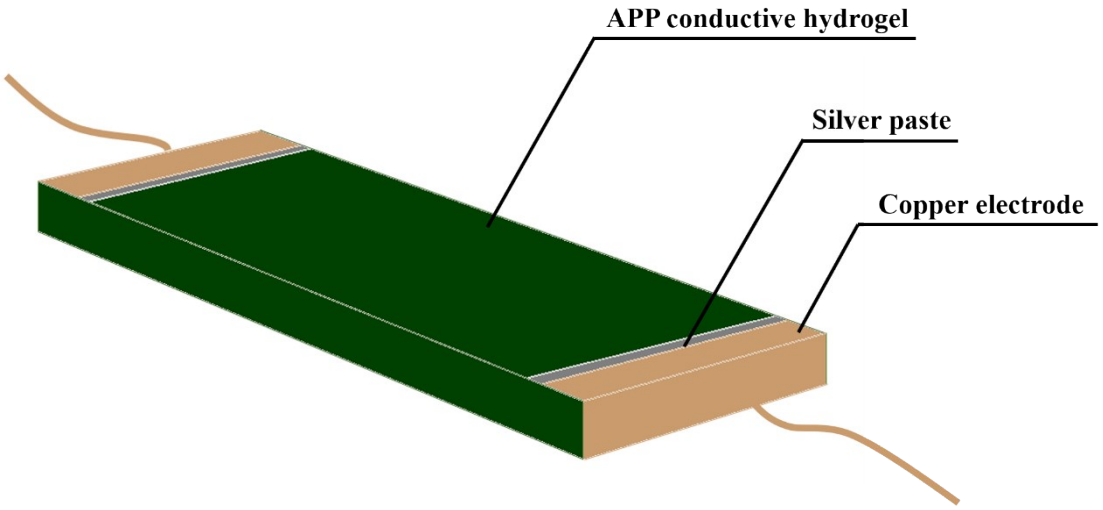

Fig. S1 Three-dimensional structure of strain sensors assembled from APP hydrogels.

Characterizations

The fracture surface morphology of freeze-dried hydrogels was observed with field emission scanning electron microscopy (FE-SEM) (FEI-QUANTA250FEG, USA). Fourier transform infrared (FTIR) spectra were recorded on a Nicolet 8700 spectrometer (Thermo Fisher Scientific, USA) at a scanning resolution of 2 cm^{-1} in the wavenumber range from 4000 to 500 cm^{-1}. Raman spectrometer (Via-H31894, Renishaw, British) were performed in the wavenumber range from 400 to 2000 cm^{-1}. X-ray diffraction (XRD) patterns were recorded on a D8-Advanced X-ray diffractometer (Bruker Co. Ltd, Switzerland) with Cu Kα radiation (λ = 0.154 nm) from 5 to 45°, at a scanning speed of 5° min^{-1}. The oscillatory frequency sweep measurements were performed at a strain amplitude of 5 % with shear frequency in the range of 0.1−100 rad/s at 25 °C to determine the storage and loss moduli of the hydrogels. The tensile process of hydrogels was characterized using a universal tensile testing machine (CMT 4254, Shenzhen SANS, China) with a tensile rate of 1 mm min^{-1} at ambient temperature. The conductivity of the hydrogel was tested by a digital source meter (Keithley 2400, Tektronix, U.S.A.), and the piezoresistive performance was also tested by it combining with a universal tensile testing machine. The digital source meter was applied to evaluate performance of APP hydrogel sensors in practical applications.

For ANFs, the characteristic bands at 3313 cm^{-1} are attributed to the N-H stretching vibrations.[2] The bands at 1631 and 1537 cm^{-1} are ascribed to the C=O stretching vibration and the N-H deformation vibration of amide group, respectively.[3] The bands at 1501 and 1309 cm^{-1} are assigned to the C=C stretching and Ph-N stretching of benzene ring, respectively.[4] The characteristic peaks of PVA are presented for the ANFs/PVA hydrogels. The peaks at 3286, 2904, and 1083 cm^{-1} are attributed to the vibrations of O-H, C-H, and C-O bonds of PVA, respectively.[5,6] In ANFs-PVA composite materials, the C=O characteristic peak of ANFs obviously moves to the low band, which proves the existence of hydrogen bond between ANFs and PVA components.[2]

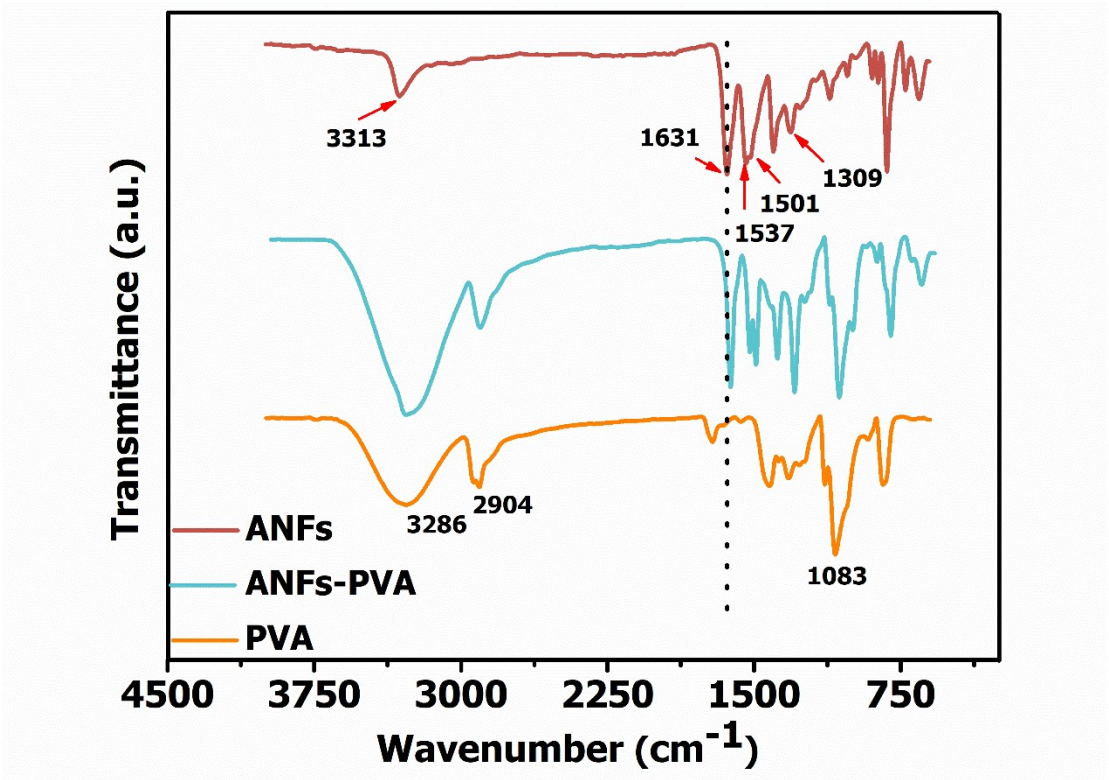

Fig. S2 FTIR spectra of ANFs, PVA and ANFs-PVA

X-ray diffraction spectroscopy (XRD) scans of PVA, ANFs, PANI and APP hydrogels are shown in Figure S3 (Supporting Information). In the X-ray diffraction curve of PVA, the crystalline peaks appear at $2\theta = 19.5°$, corresponding to the (101) plane of the PVA.[6] The ANFs shows three typical Kevlar characteristic peaks, which

can be assigned to the (110), (200), and (004) diffractions, respectively.[2] For the PANI, two broad peaks at around 2θ = 20.6 and 25.1° are ascribed to the periodicity parallel and perpendicular to PANI chains.[3] In the case of APP, the main characteristic peak was consistent with ANFs, but the intensity decreased significantly, which may be attributed to the introduction of the PVA and PANI.[4]

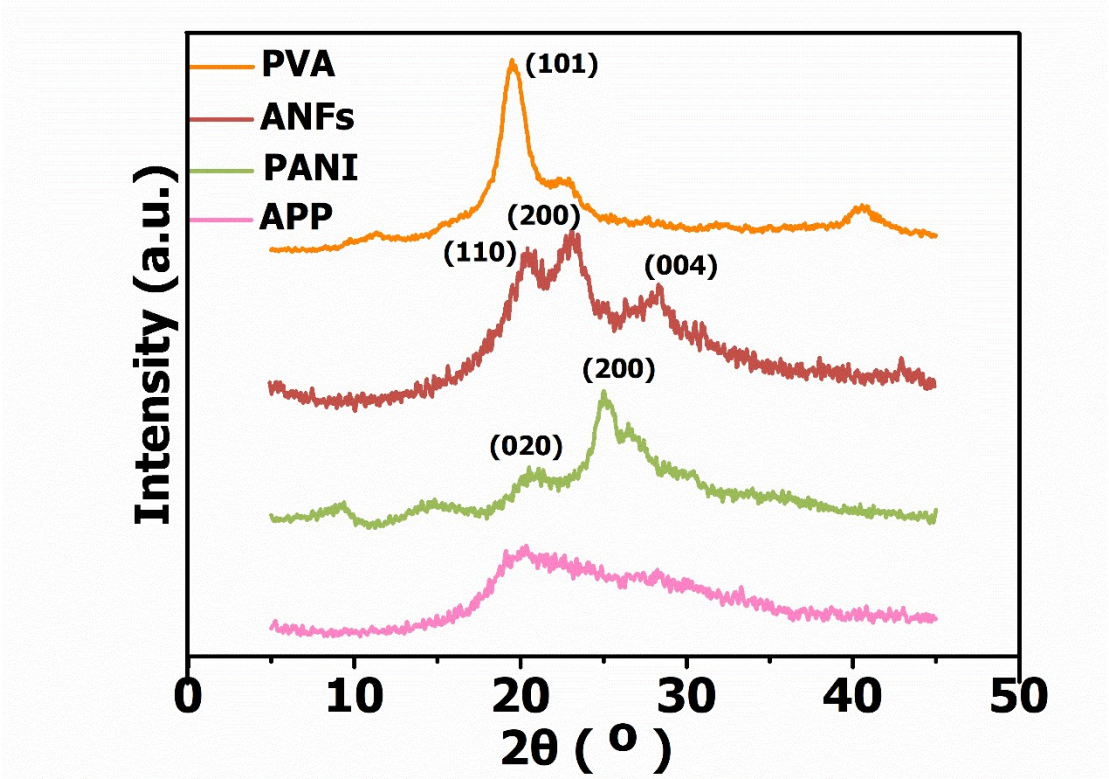

Fig. S3 XRD spectra of PVA, ANFs, PANI and APP.

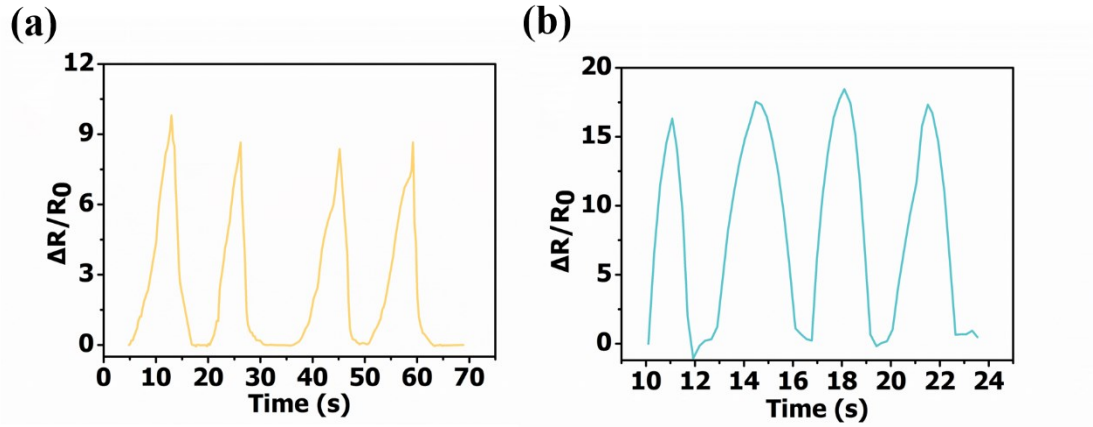

Fig. S4 (a) The stretching cycle at 60% strain. (b) The stretching cycle at 100% strain.

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